Geochemistry Geophysics Geosystems G3

*Article*
Volume 14, Number 5
17 May 2013
doi:10.1002/ggge.20116
ISSN: 1525-2027

Published by AGU and the Geochemical Society

# Asian dust input in the western Philippine Sea: Evidence from radiogenic Sr and Nd isotopes

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[1] The radiogenic strontium (Sr) and neodymium (Nd) isotope compositions of the detrital fraction of surface and subsurface sediments have been determined to trace sediment provenance and contributions from Asian dust off the east coast of Luzon Islands in the western Philippine Sea. The Sr and Nd isotope compositions have been very homogenous near the east coast of the Luzon Islands during the latest Quaternary yielding relatively least radiogenic Sr ($^{87}Sr/^{86}Sr = 0.70453$ to 0.70491) and more radiogenic Nd isotope compositions ($\varepsilon_{Nd}(0) = +5.3$ to +5.5). These isotope compositions are similar to Luzon rocks and show that these sediments were mainly derived from the Luzon Islands. In contrast, the Sr and Nd isotope compositions of sediments on the Benham Rise and in the Philippine Basin are markedly different in that they are characterized by overall more variable and more radiogenic Sr isotope compositions ($^{87}Sr/^{86}Sr = 0.70452$ to 0.70723) and less radiogenic Nd isotope compositions ($\varepsilon_{Nd}(0) = -5.3$ to +2.4). The Sr isotope composition in the Huatung Basin is intermediate between those of the east coast of Luzon and Benham Rise, but shows the least radiogenic Nd isotope compositions. The data are consistent with a two end-member mixing relationship between Luzon volcanic rocks and eolian dust from the Asian continent, which is characterized by highly radiogenic Sr and unradiogenic Nd isotope compositions. The results show that Asian continental dust contributes about 10–50% of the detrital fraction of the sediments on Benham Rise in the western Philippine Sea, which offers the potentials to reconstruct the climatic evolution of eastern Asia from these sediments and compare this information to the records from the central and northern Pacific.

**Components:** 9,200 words, 5 figures, 1 table.

**Keywords:** Asian dust; Luzon Islands; detrital fraction; Sr isotope; Nd isotope; western Philippine Sea.

**Index Terms:** 0305 Atmospheric composition and structure: Aerosols and particles (0345, 4801, 4906); 1040 Geochemistry: Radiogenic isotope geochemistry.

**Received** 20 December 2012; **Revised** 12 March 2013; **Accepted** 12 March 2013; **Published** 17 May 2013.

Jiang, F., M. Frank, T. Li, T.-Y. Chen, Z. Xu, and A. Li (2013), Asian dust input in the western Philippine Sea: Evidence from radiogenic Sr and Nd isotopes, *Geochem. Geophys. Geosyst.*, *14*, 1538–1551, doi:10.1002/ggge.20116.

## 1. Introduction

[2] Eolian dust plays a significant role in climate change through either direct or indirect interactions with radiation [e.g., *Tegen et al.*, 1996; *Ridgwell et al.*, 2002] or ocean biogeochemistry [e.g., *Martin*, 1990]. At the same time, eolian dust records provide valuable insights into changes of atmospheric conditions and climatic changes on land in the geological past [*Kohfeld and Harrison*, 2001]. At present, the Asian continent is the second largest dust source area on the earth [*Han et al.*, 2008] and contributes large amounts of terrigenous material to the Pacific Ocean (about 400 Tg/yr, *Han et al.*, [2011]). Radiogenic Sr, Nd, and Pb isotope evidence indicates that the eolian dust derived from Asian continent has been widely distributed across the mid-latitude Pacific [*Nakai et al.*, 1993; *Jones et al.*, 1994; 2000; *Rea*, 1994; *Mahoney et al.*, 1998; *Pettke et al.*, 2000]. Mass accumulation rate and grain size variation of Asian dust in central North Pacific sediment have been used to trace and reconstruct the atmospheric circulation and paleoclimatic conditions of the Asian continent [*Nakai et al.*, 1993; *Rea*, 1994; *Asahara et al.*, 1999; *Asahara*, 1999]. The tropical northwestern Pacific has been a key area influencing and modulating global climate change by serving as one of the main heat engines of global climate and as a vapor source for the global hydrological cycle [*Lea*, 2002]. However, eolian dust deposition in the low latitude tropical northwestern Pacific has not been studied in much detail as yet.

[3] The western Philippine Sea is the largest back-arc basin in the low latitude northwestern Pacific Ocean. It is bounded by the Ryukyu, Philippine trench and East Luzon Trench/Island arc systems in the west and the Palau-Kyushu Ridge in the east (Figure 1). The North Equatorial Current (NEC) bifurcates as it approaches the east bank of the Philippines splitting into the northward flowing Kuroshio Current (KC) and the southward flowing Mindanao Current [e.g., *Toole et al.*, 1990]. The KC flows north along the east coast of Taiwan with no obvious seasonal variation. The estimated depth of the KC varies from a few hundred to 1000 m. The predominant wind system in this area during the late Quaternary has been the East Asia monsoon [*An*, 2000], whereby the northeasterly monsoon prevails from November to March [*Chuang and Liang*, 1994].

[4] The area of this study is the western Philippine Sea and the Benham Rise, which is a mid-plate rise at the westernmost edge of the western Philippine Basin, and mostly shallower than 2900 m. Previous studies showed that the Late Quaternary sediments on Benham Rise are mainly composed of calcareous nannofossil ooze and foraminifera, and volcanic debris [*The Shipboard Scientific Party*, 1975; *Wei et al.*, 1998]. Volcanic debris (brown and clear volcanic glass, plagioclase, amphibole, and rare rock fragments) is almost ubiquitous and enriched in some layers corresponding to major volcanic eruptions of Luzon Island [*Wei et al.*, 1998; *Ku et al.*, 2009]. Terrigenous detrital material appears to be virtually absent and is represented only by minor amounts of pelagic clay indicating that Benham Rise was shielded from supply of terrigenous detritus from the Luzon Island and other continental landmasses to the west. *Ren et al.* [2007] argued that seamounts consisting of intermediate acidic rock may represent the main origin of felsic debris, whereas terrigenous matter from China's mainland and the Philippine Island arc have had little quantitative influence on the sedimentation on Benham Rise and western Philippine Basin. On the contrary, on the basis of decreasing deposition rates towards the open Pacific, *Huh et al.* [1992] suggested that the main source of material in western Philippine Sea has been the Asian continent and that eolian transport has been the primary pathway of terrestrial input to this area despite the fact that precipitation rates have been very high. In support of this, loess-like sediment layers have been found in several short sediment cores on the east coast of Luzon Islands, and studies of the mineral compositions and grain size indicated that these sediments were derived from the Asian continent [*Qin et al.*, 1995; *Shi et al.*, 1994, 1995; *Chen et al.*, 1997]. However, the temporal and spatial distribution of the loess-like sediments is very limited. Consequently, whether terrigenous detritus or eolian dust from the Asian continent has been carried to the western Philippine Sea, and how widespread it is, is not resolved.

[5] The radiogenic isotope compositions of Sr and Nd are powerful and sensitive proxies to characterize marine sediment provenance [*Grousset et al.*, 1988; *Parra et al.*, 1997; *Biscaye et al.*, 1997; *Mahoney et al.*, 1998; *Asahara et al.*, 1999; *Yokoo et al.*, 2004; *Chen et al.*, 2007; *Yang et al.*, 2007; *Dou et al.*, 2012]. While the Sr isotope composition can, to some extent, be influenced by grain-sized sorting and diagenesis of the sediment [*Dasch*, 1969; *Walter et al.*, 2000], the Nd isotope composition retains the information on sediment source

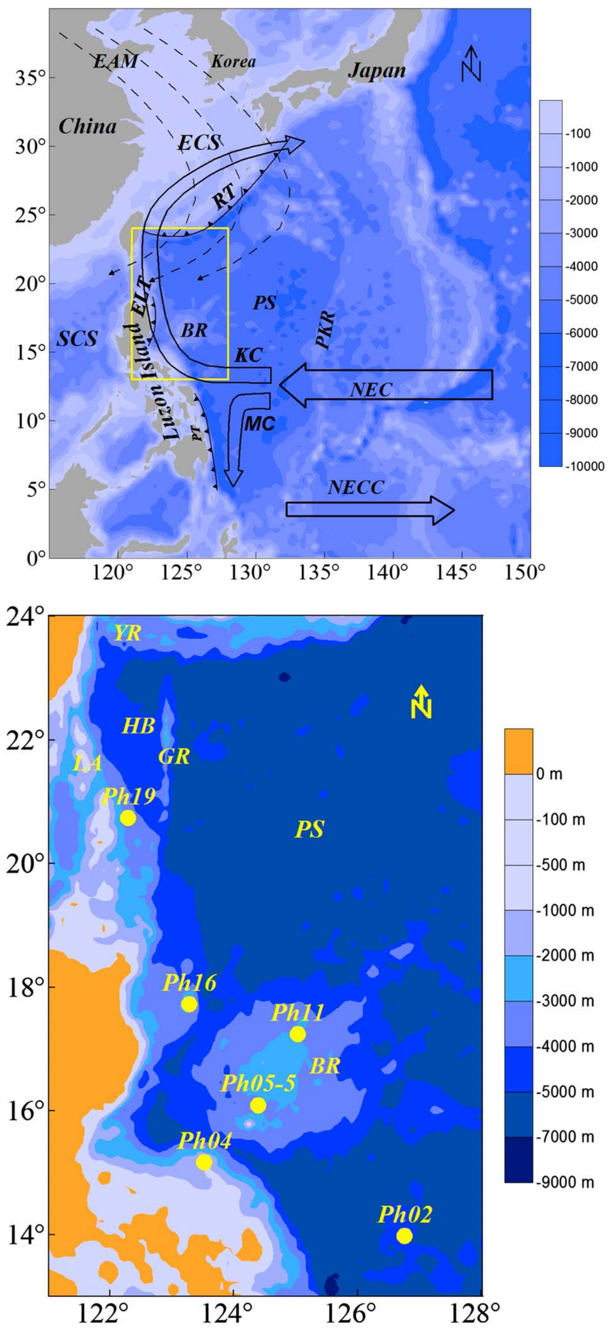


**Figure 1.** Location of study area (The inset map represents the enlargement of the yellow frame. The present day flow paths of the main currents are reported by black arrows, NEC-North Equatorial Current, KC-Kuroshio Current, MC-Mindanao Current, NECC-North Equatorial Counter Current. The black dashed lines indicate the prevailing air trajectories of the East Asian Monsoon (EAM). ECS-East China Sea, SCS-South China Sea, BR-Benham Rise, PS-Philippine Sea, YR-Yaeyama Ridge, LA-Luzon Arc, HB-Huatung Basin, GR-Gagua Ridge, PKR-Palau-Kyushu Ridge, RT-Ryukyu Trench, ELT-East Luzon Trench, PT-Philippine Trench).

area during weathering, transport, and submarine sediment redistribution processes [*Goldstein et al.*, 1984; *Walter et al.*, 2000]. The combination of Sr and Nd isotope signatures has been shown to reliably discriminate inputs from different detrital sediment source areas [*Revel et al.*, 1996; *Biscaye et al.*, 1997; *Weldeab et al.*, 2002].

[6] In western Pacific, different crustal domains have different Sr and Nd isotopic composition, and the detrital sediments in the basins adjacent to these source rock domains consequently have signatures characteristic of these domains [*Mahoney et al.*, 1998]. For the eastern Philippine Sea basin, some studies have shown that the Sr and Nd isotopic signature of the sediments primarily represents variable degrees of mixing between detritus from old Asian continental dust (more evolved Sr and Nd isotope signatures) and juvenile Pacific rim volcanics (juvenile Sr and Nd isotope signatures), and thus recorded the evolution of sediment provenance in this area [*Mahoney et al.*, 1998; *Asahara et al.*, 1995].

[7] In this study, we evaluate the importance of eolian detrital input in the western Philippine Sea by comparing the Sr and Nd isotope composition of the detrital sediments to potential source areas, such as the Luzon Island, Eastern Taiwan, and eolian dust in the western North Pacific.

## 2. Material and Methods

### 2.1. Material

[8] A total of 30 samples from six sediment cores (four to six samples of the uppermost 102 cm for each core) were analyzed covering the east coast of the Luzon Islands (Ph04), Benham Rise (Ph05-5 and Ph11), the western Philippine Basin (Ph02 and Ph16), and the Huatung Basin (Ph19) (Figure 1 and Table 1). The sediments were recovered during a cruise with RV Ke Xue Yi Hao in 2004. According to the nannofossil biochronology and oxygen isotope age models for core MD97-2143 [*Wei et al.*, 1998; *Horng et al.*, 2002] and AMS$^{14}$C datings of core Ph05-5 [*Li et al.*, 2010] on Benham Rise, the sedimentation rates of the top 102 cm of sediments in this area is about 2–3 cm/ka, which implies that the depositional age of the studied samples covers up to the past 50 kyr and that the deposition rate of the detrital fraction in this area has ranged from 0.4 to 2.7 g cm$^{-2}$ kyr$^{-1}$ (*Jiang et al.*, unpublished data).

**Table 1.** The Analyzed Samples and Sr, Nd Isotopic Compositions of Detrital Fraction in Sediments of the Western Philippine Sea

| Location | Depth (cm) | $^{87}Sr/^{86}Sr$ | ±1 S.E.($10^{-6}$) | $^{143}Nd/^{144}Nd$ | ±1 S.E.($10^{-6}$) | εNd(0) |
|---|---|---|---|---|---|---|
| Philippine Basin | Ph02: Latitude: 13°58.99′N–Longitude: 126°44.69′E–Water Depth: 4432 m | | | | | |
| | 0–2 | 0.70626 | 6 | 0.512551 | 3 | −1.7 |
| | 10–12 | 0.70616 | 5 | 0.512525 | 4 | −2.2 |
| | 20–22 | 0.70487 | 6 | 0.512666 | 4 | 0.5 |
| | 36–38 | 0.70720 | 8 | 0.512438 | 4 | −3.9 |
| | 50–52 | 0.70723 | 6 | 0.512470 | 3 | −3.3 |
| | Ph16: Latitude: 17°42.63′N – Longitude: 123°16.34′E – Water Depth: 2500 m | | | | | |
| | 0–3 | 0.70485 | 6 | 0.512760 | 5 | 2.4 |
| | 10–12 | 0.70459 | 6 | 0.512718 | 7 | 1.6 |
| | 20–22 | 0.70491 | 6 | 0.512691 | 4 | 1.0 |
| | 30–32 | 0.70461 | 7 | 0.512709 | 7 | 1.4 |
| | 50–52 | 0.70452 | 6 | 0.512714 | 6 | 1.5 |
| | 100–102 | 0.70488 | 5 | 0.512689 | 5 | 1.0 |
| Benham Rise | Ph05-5: Latitude: 16°02.96′N – Longitude: 124°20.69′E – Water Depth: 3382 m | | | | | |
| | 4–6 | 0.70625 | 6 | 0.512647 | 4 | 0.2 |
| | 12–14 | 0.70644 | 7 | 0.512634 | 4 | −0.1 |
| | 28–30 | 0.70662 | 7 | 0.512531 | 6 | −2.1 |
| | 52–54 | 0.70636 | 6 | 0.512503 | 4 | −2.6 |
| | 68–70 | 0.70669 | 7 | 0.512470 | 4 | −3.3 |
| | Ph11: Latitude: 17°13.52′N – Longitude: 125°00.47′E – Water Depth: 2812 m | | | | | |
| | 2–4 | 0.70638 | 6 | 0.512601 | 5 | −0.7 |
| | 11–13 | 0.70636 | 8 | 0.512481 | 8 | −3.1 |
| | 20–22 | 0.70651 | 7 | 0.512495 | 4 | −2.8 |
| | 52–54 | 0.70607 | 6 | 0.512543 | 6 | −1.9 |
| East coast of Luzon | Ph04: Latitude: 14°52.12′N – Longitude: 123°29.60′E – Water Depth: 772 m | | | | | |
| | 0–3 | 0.70464 | 7 | 0.512916 | 4 | 5.4 |
| | 6–8 | 0.70455 | 7 | 0.512916 | 5 | 5.4 |
| | 10–12 | 0.70453 | 8 | 0.512912 | 4 | 5.4 |
| | 22–24 | 0.70461 | 7 | 0.512910 | 3 | 5.3 |
| | 30–32 | 0.70462 | 6 | 0.512919 | 4 | 5.5 |
| Huatung Basin | Ph19: Latitude: 20°26.03′N – Longitude: 122°28.54′E – Water Depth: 2970 m | | | | | |
| | 2–4 | 0.70573 | 5 | 0.512407 | 7 | −4.5 |
| | 10–12 | 0.70542 | 6 | 0.512378 | 4 | −5.1 |
| | 22–24 | 0.70608 | 6 | 0.512370 | 4 | −5.2 |
| | 30–32 | 0.70599 | 6 | 0.512365 | 6 | −5.3 |
| | 42–44 | 0.70471 | 6 | 0.512560 | 7 | −1.5 |

## 2.2. Analytical Methods

[9] Approximately 2 g of bulk sediment of each sample was dried (at 50°C) and homogenized in an agate mortar. In order to determine pure detrital radiogenic Sr and Nd isotope signatures, we use the leaching procedure following the method applied in *Gutjahr et al.* [2007] and *Bayon et al.* [2002]. Sample splits of 300–400 mg each were treated with distilled water, acetic acid (10%), hydroxylamine-hydrochloride (0.05 M), and hydrogen peroxide (5%) at room temperature to remove sea salts, carbonates, ferromanganese coatings, and organic matter, respectively. Subsequently, the residual detrital fractions were dried (at 50°C) and ground again. About 50 mg of each sample was used for total dissolution in a mixture of concentrated HCl, $HNO_3$, and HF on a hot plate. In order to document the validity of this dissolution method in the study area, we performed duplicate analyses of the same samples, as well as a comparison between complete (no more mineral grains visible) and incomplete hot plate dissolutions for selected samples, which were

in all cases identical within analytical uncertainties (Appendix 1 and 2 in the supporting information)[1]. Sr and Nd were separated and purified for mass spectrometric analyses by application of standard ion chromatographic procedures [*Cohen et al.*, 1988; *Horwitz et al.*, 1992; *Lugmair and Galer*, 1992].

[10] Radiogenic Sr and Nd isotope measurements were performed on a Nu Instruments Multicollector Inductively Coupled Plasma Mass Spectrometer at GEOMAR Kiel. The measured $^{87}Sr/^{86}Sr$ ratios were interference ($^{86}Kr$, $^{87}Rb$) and mass bias corrected (using $^{86}Sr/^{88}Sr = 0.1194$, *Steiger and Jäger*, [1977]). The Sr isotope results were normalized to NBS987 $^{87}Sr/^{86}Sr = 0.71025$. Repeated standard measurements gave an external reproducibility better than 43 ppm (2σ). Measured Nd isotopic compositions were corrected for instrumental mass bias using a $^{146}Nd/^{144}Nd$ of 0.7219. External reproducibility was estimated by repeated measurements of the JNdi-1 standard and was always better than 65 ppm (2σ). $^{143}Nd/^{144}Nd$ ratios were normalized to the accepted value for JNdi-1 of 0.512115 [*Tanaka et al.*, 2000]. For convenience, $^{143}Nd/^{144}Nd$ ratios are expressed as $\varepsilon_{Nd}(0) = (^{143}Nd/^{144}Nd/0.512638 - 1) \times 10^4$ [*Jacobsen and Wasserburg*, 1980].

[11] The National Oceanic and Atmospheric Administration (NOAA) Hybrid Single-Particle Lagrangian Integrated Trajectory (HYSPLIT) model [*Draxler and Rolph*, 2012; *Rolph*, 2012] was used to calculate three-dimensional backward trajectories of the dust particles during one dust event (from 27 March to 1 April 2006, *Liu et al.*, [2009]) to constrain the source area of the detrital fraction in the sediment of western Philippine Sea. Air mass back trajectories for arrival heights of 100, 500, and 1000 m above the western Philippine Sea (e.g., site Ph05-5 and site Ph02), together with Taipei [*Liu et al.*, 2009] during this dust event were obtained.

## 3. Results

[12] Sr and Nd isotope data for the samples of our study are graphically displayed as $^{87}Sr/^{86}Sr$ versus $\varepsilon_{Nd}(0)$ on Figure 2 and are provided in Table 1. The Sr and Nd isotope compositions of all samples are significantly less radiogenic in Sr and more radiogenic in Nd than Asian dust [*Pettke et al.*, 2000]. There are significant differences between the different locations as a consequence of the nearby geological framework of the rocks on land. More juvenile Sr and Nd isotope ratios are observed in samples on the east coast of the Luzon Islands (Ph04). The range of Sr isotope signatures is very narrow and similar to Luzon rocks [*Defant et al.*, 1990]. More evolved Sr (Nd) and more variable Sr isotope ratios (>0.7045) are characteristic for samples from Benham Rise (Ph05-5, Ph11) and in the western Philippine Basin (Ph02, Ph16). The Sr and Nd isotope signature of the Huatung Basin (Ph19) is distinct from Benham Rise, the western Philippine Sea, and east coast of Luzon Islands in that it shows the most evolved Nd isotope signatures, whereas the Sr isotope signatures are more evolved than those of the east coast of Luzon but less evolved than on Benham Rise and in the Philippine Basin (Figure 2).

## 4. Discussion

### 4.1. Potential Mixing End-Members of the Sediments

[13] In order to identify the potential sediment source of the study area, the published $^{87}Sr/^{86}Sr$ and/or $^{143}Nd/^{144}Nd$ ratios (expressed as $\varepsilon_{Nd}(0)$) of sediments and/or rocks of the study area and the detrital fractions of the sediments in the western Philippine Sea (this study) were plotted in Figure 3.

[14] Old Asian continentally derived materials, such as Chinese loess [*Chen et al.*, 2007] and sediment in East China Sea [*Asahara et al.*, 1995], sediment from eastern Taiwan Orogen and Huatung Basin [*Bentahila et al.*, 2008], and marine sediments in western and central North Pacific [*Asahara et al.*, 1999; *Jones et al.*, 1994; *Pettke et al.*, 2000; *Mahoney*, 2005] are more radiogenic in their Sr isotope composition ($^{87}Sr/^{86}Sr$ ratios >0.71000) and less radiogenic in Nd isotope compositions (e.g., $\varepsilon_{Nd}(0) < -9$ in Chinese loess). In contrast, the relatively young crustal province around the Pacific Rim, such as Luzon Islands [*Defant et al.*, 1990], is characterized by sediment with homogeneous, less radiogenic Sr, and highly radiogenic Nd isotope compositions (with $^{87}Sr/^{86}Sr$ ratios <0.705, $\varepsilon_{Nd}(0) > +5$). The Sr and Nd isotope signatures of the sediments in the marginal basins of the western Pacific are intermediate between the older continental signatures and those of the young volcanic arc (Figure 3) and are characterized by more variable Sr and Nd isotope compositions.

[1]All supporting information can be found in the online version of this article.

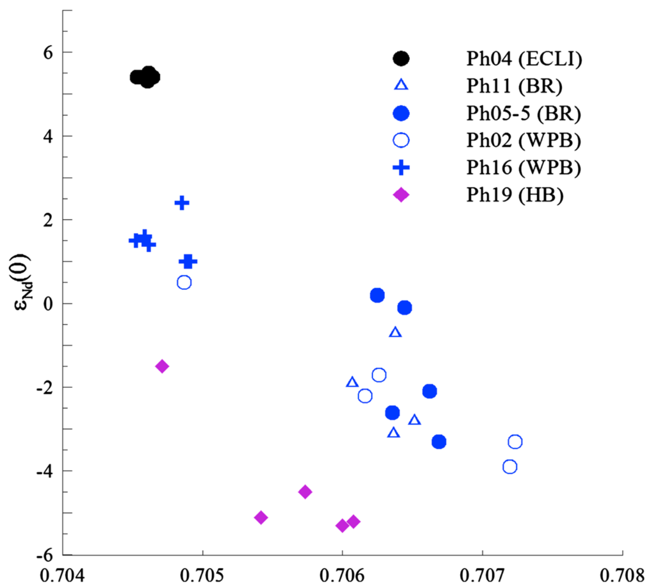


**Figure 2.** $^{87}Sr/^{86}Sr$ vs. $\varepsilon_{Nd}(0)$ of the detrital fraction of surface and subsurface sediment from the East Coast of the Luzon Islands (ECLI), the Benham Rise (BR), the Western Philippine Basin (WPB), and the Huatung Basin (HB).

[15] The Sr isotope compositions of the detrital fraction in sediment on Benham Rise and in the western Philippine Basin are more radiogenic (0.70452–0.70723) than those of the sediments east of coastal Luzon Islands (0.7045–0.7046), the rocks of the Luzon Islands [*Defant et al.*, 1990], and volcanic glasses in the sediment of Benham Rise, which are also derived from the Luzon Islands [*Ku et al.*, 2009]. Obviously, the Luzon Islands contribute detrital material with least radiogenic Sr isotope compositions to Benham Rise sediments. In contrast, the more radiogenic Sr isotope compositions of Benham Rise and the western Philippine Basin sediments require an extra-basinal sediment source and/or geological processes (e.g., weathering, grain-size effects, etc.). According to the distribution of $^{87}Sr/^{86}Sr$ ratios and of $\varepsilon_{Nd}(0)$ signatures (Figure 3), the older continental crustal areas around the western Philippine Sea, including eastern Taiwan and the Asian continent have been potential contributors of detrital material with evolved Sr and Nd isotope signatures to the Benham Rise and the western Philippine Basin.

## 4.2. Luzon Islands: Potential Source Area for Sediments on Benham Rise?

[16] The particulate Sr isotope composition in marine sediments is mainly controlled by weathering of different rocks [*Blum and Erel*, 1995]. Chemical and Sr isotope analyses of the tephra layers and the deep-sea tephrostratigraphic record of core MD97-2143 from the east side of Luzon Island (Philippines) have been used to infer that the volcanic material deposited on Benham Rise has originated from the Macolod Corridor in the southwestern part of the Luzon for the last 1.9 Myr [*Ku et al.*, 2009]. The $^{87}Sr/^{86}Sr$ ratios of these particles range from 0.70384 to 0.70479 in the tephra layers [*Ku et al.*, 2009]. The $^{87}Sr/^{86}Sr$ ratios of detrital fraction in sediments near the east coast of the Luzon Islands (Ph04) range from 0.70453 to 0.70464, which is close to that of the volcanic rocks of the Luzon Islands ranging from 0.703 to 0.704 [*Defant et al.*, 1990]. Thus, the relatively radiogenic Sr isotope compositions of the detrital fraction on Benham Rise cannot originate from erosion of the Luzon Islands.

## 4.3. Grain-Size Effects on Sr and Nd Isotope Compositions

[17] Grain-size has been demonstrated to influence the Sr isotope ratios of detrital sediments due to varying mineralogical and hence isotopic composition of different size fractions. The fine-grained fraction tends to be enriched in radiogenic $^{87}Sr$ as a consequence of high Rb/Sr in micas and biotite [*Dasch*, 1969; *Grousset et al.*, 1992; *Feng et al.*, 2009]. Therefore, the $^{87}Sr/^{86}Sr$ ratio alone is an ambiguous indicator of sediment provenance. In contrast, size fractions from the same source have similar $\varepsilon_{Nd}(0)$ values which allow for a reconstruction of particle provenance in deep-sea sediments [*Tütken et al.*, 2002]. Studies on the Nd isotope composition of sediment with different grain-sizes in river system [*Goldstein et al.*, 1984], eolian dust [*Nakai et al*,

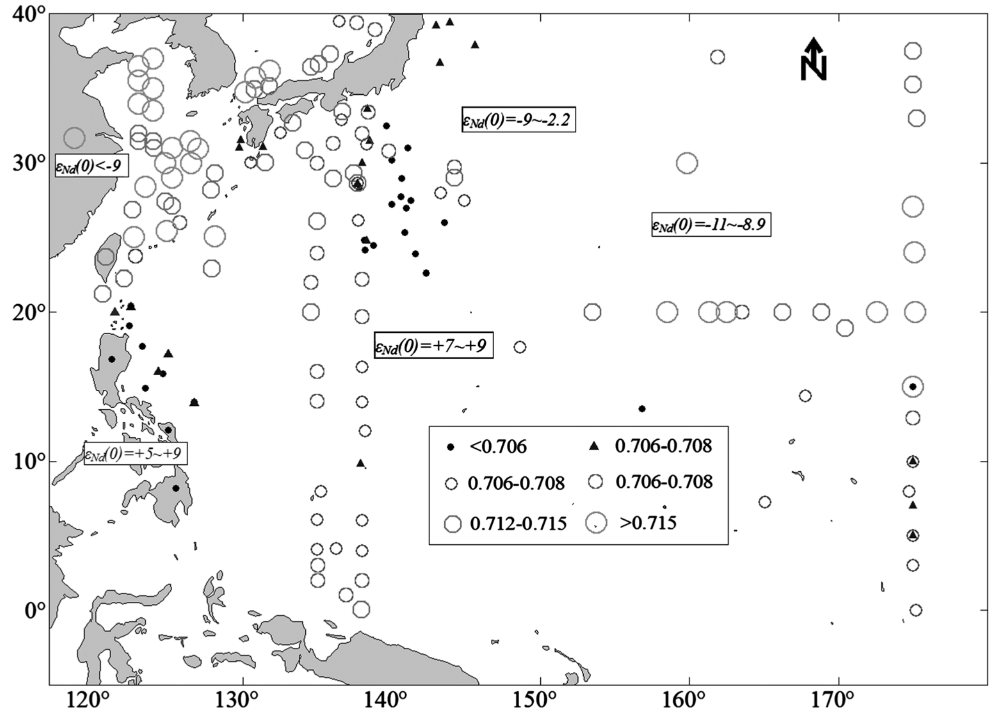


**Figure 3.** $^{87}Sr/^{86}Sr$ ratios and $\varepsilon_{Nd}(0)$ of detrital fraction of surface and subsurface sediment from study area (ref. Figure 1, this study), and the potential source sediments [*Jones et al.*, 2000; *Pettke et al.*, 2000; *Bentahila et al.*, 2008, *Asahara et al.*, 1995, 1999; *Mahoney*, 2005; *Woodhead*, 1989; *Ku et al.*, 2009; *Page and Johnson*, 1974, *Smith and Compston*, 1982].

1993; *Chen et al.*, 2013], and marine sediment [*Walter et al.*, 2000; *Tütken et al.*, 2002] confirmed that the Nd isotope composition is nearly independent of the grain-size of the sediment fractions. The Nd isotope compositions of volcanic material from the Luzon Islands are homogeneous and most of the $\varepsilon_{Nd}(0)$ values are above +5 [*Defant et al.*, 1990]. The $\varepsilon_{Nd}(0)$ values of sediments from off the east coast of Luzon (Ph04, Table 1 and Figure 4) are between +5.3 and +5.5, similar to that the volcanic material from Luzon. In contrast, the Nd isotope ratios in Benham Rise (Ph05-5 and Ph11) and Philippine Basin sediments (Ph02 and Ph16) are less radiogenic ($\varepsilon_{Nd}(0) = +2.4$ to $-4$), which is clearly not a consequence of grain size fractionation. Thus, the large Sr isotope variations are also inferred to primarily reflect the original source rocks, which is consistent with the young age of these rocks that did not allow for significant ingrowth of radiogenic $^{87}Sr$ in the micas. The unradiogenic $\varepsilon_{Nd}(0)$ values and radiogenic Sr isotope ratios found in the western Philippine Sea sediments thus require an extrabasinal sediment source.

## 4.4. Is the Taiwan Orogen a Possible Contributor of Sediments to Benham Rise?

[18] The Taiwan orogen is situated at the eastern margin of the Eurasian continental plate, and mainly composed of Cenozoic continental shelf and slope deposits lying unconformably on a late Paleozoic metamorphic basement and the Miocene–Pleistocene arc-related Coastal Range. The Taiwan orogen is characterized by very high sediment discharge because of the ongoing arc-continent collision and related uplift combined with very high precipitation and intense weathering. The erosion rates in the eastern central range have been on the order of 3–6 mm yr$^{-1}$ over at least the entire Quaternary [*Dadson et al.*, 2003]. The eastern Taiwan orogen is thus a potential source area of sediment in the Philippine Sea. However, the results of our Sr and Nd isotope study do not support this.

[19] If we consider northern Luzon Islands as the juvenile Sr and Nd end-member and eastern Taiwan material as the evolved Sr and Nd end-member, a mixing curve can be calculated. If the sediments on Benham Rise and in the western Philippine Basin were indeed a mixture of eastern Taiwan and Luzon Island inputs, they should plot on this mixing curve. For this consideration, the choice of the exact end-member values of Sr and Nd concentration and isotopic compositions is crucial. The Sr and Nd isotope value of the northern Luzon Islands is well defined by the homogeneous isotopic compositions of their rocks [*Defant et al.*,

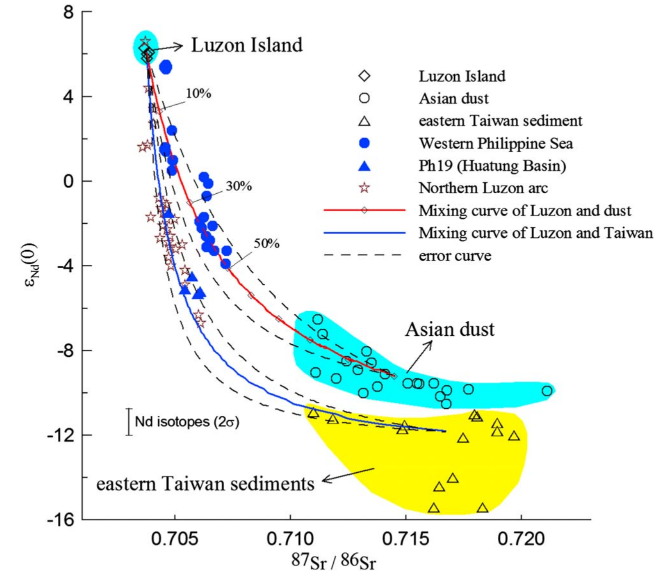


**Figure 4.** $^{87}Sr/^{86}Sr$ versus $\varepsilon_{Nd}(0)$ of sediments and rocks of potential source areas (literature data) and sediments from Philippine Sea (this study) (The red line is the mixing curve of volcanic material of the Luzon Islands and eolian dust as encountered in the North Pacific, and the relative contents of Asian dust are marked on the line. The blue line is the mixing curve of volcanic material from the Luzon Islands and erosion products of the eastern Taiwan orogen. The error bars of Sr isotope are smaller than symbol size. The concentrations of Sr and Nd and isotopic compositions used here are Sr = 214.1 ppm, Nd = 38ppm, $^{87}Sr/^{86}Sr$ = 0.71449, and $^{143}Nd/^{144}Nd$ = 0.51217 for average eolian dust in the North Pacific (*Pettke et al*., 2000); Sr = 440 ppm, Nd = 19 ppm, $^{87}Sr/^{86}Sr$ = 0.70376, and $^{143}Nd/^{144}Nd$ = 0.51295 for average volcanic material from the Luzon Islands [*Defant et al*., 1990]; Sr = 58 ppm, Nd = 31 ppm, $^{87}Sr/^{86}Sr$ = 0.71669, and $^{143}Nd/^{144}Nd$ = 0.51203 for the average composition of the eastern Taiwan orogen [*Chen et al*., 1990; *Chen and Lee*, 1990; *Bentahila et al*., 2008]; The Sr and Nd isotope data of Northern Luzon Arc are from *Defant et al*. [1990] and *Vidal et al*. [1989]).

1990]. In contrast, the Sr and Nd isotopic signature of eastern Taiwan material is less well constrained and homogenous. Published Sr and Nd isotope compositions of major rock formations [*Chen et al*., 1990] and of sediments of major rivers of eastern Taiwan [*Chen and Lee*, 1990] indicate that the Sr (Nd) isotope compositions of the Taiwanese rocks and sediments are overall significantly more radiogenic (less radiogenic) [*Chen et al*., 1990; *Lan et al*., 1995, 2002; *Bentahila et al*., 2008] than those of the western Philippine Basin, Benham Rise, and Luzon Island. However, the range of Sr and Nd isotope compositions of Taiwanese rocks and sediments is very large, e.g., $^{87}Sr/^{86}Sr$ range from 0.70531 to 0.72216 [*Bentahila et al*., 2008], which makes it difficult to identify an unambiguous end-member composition for the western Philippine Sea sediments.

[20] *Chen and Lee* [1990] analyzed the Sr and Nd isotope of sediments from the beds of some major Taiwanese River sediment and found that they are composed predominantly of continent-derived detritus that is in contrast to other arc-related regions such as Luzon where river sediments reflect mostly their island and continental arc igneous source lithologies [*Chen and Lee*, 1990]. The Sr and Nd isotope composition of the river sediments are similar to, but more homogeneous than those observed for sedimentary formations in their source areas. An Nd isotope study indicates that the river sediments of southeast coastal range (e.g., Peniantahsi and Hsuikuluanhsi rivers), from where most of the sediment to the Huatung Basin is supplied [*Dadson et al*., 2003], reflect only the characteristics of Cenozoic and Late Paleozoic continental-derived sedimentary and meta-sedimentary sequences with little input of volcanic and/or phiolitic materials despite the evident occurrences of mantle-derived outcrops in their drainage areas, which resemble the sediments from the rivers without evident mantle-derived materials in their drainage areas (such as Lanyanghsi, Tsengwenhsi, and Kaopinghsi rivers).

[21] An isotope geochemical study of the Miocene to Late Paleozoic sediments and metasediments of Central and Southern Cross Island Highways from Taiwan show that the average Sr and Nd isotopic compositions of the sediments ($^{87}Sr/^{86}Sr$ = 0.71669, $\varepsilon_{Nd}(0) = -11.9$) resemble the average river sediments ($^{87}Sr/^{86}Sr$ = 0.71522, $\varepsilon_{Nd}(0) = -11.4$) reported by *Chen and Lee* [1990] indicating their same origin.

Based on the relief and erosion model of Taiwan orogen [*Dadson et al.*, 2003], we argue that the sediments from the Southern Cross Island Highways represent the typical signature of the eastern Taiwan orogen, and the Sr and Nd isotopic compositions of sediment from this regime reported by *Chen et al.* [1990] represent the eastern Taiwan end-member (The Paleozoic sediment data are excluded because *Chen et al.* [1990] argued that the radiogenic Nd isotope signature may have been influenced by the addition of juvenile mantle components). As *Chen et al.* [1990] did not report the Sr concentration of Taiwan sediments, we use here the average Sr concentration of the Peinan River suspended loads (57.8 ppm) measured by *Bentahila et al.* [2008], which represents a large-scale integrated samples of the present day Taiwan eroded sediments.

[22] Using these Sr and Nd concentrations and isotopic compositions as the end-member values, the mixing curve of eastern Taiwan and Luzon Islands can be calculated (Figure 4) and does not pass through the Benham Rise and western Philippine Basin data but through those of the northern Luzon arc rocks [*Defant et al.*, 1990; *Vidal et al.*, 1989] and Huatung sediments (Ph19). This suggests that the detrital fraction of Benham Rise sediments does not consist of a mixture of material from the northern Luzon and eastern Taiwan. Instead, the northern Luzon arc rock and Huatung sediments are consistent with a mixture of these two end-members. This result is also in agreement with the Sr-Pb isotope evidence of sediment in Huatung Basin and Luzon arc [*Bentahila et al.*, 2008], and results of the sedimentary, environmental, and tectonic evolution of the southern Huatung Basin [*Hung*, 2010].

[23] This conclusion is supported by additional geological evidence. Most of the sediment of the eastern coastal range of the Taiwan orogen is delivered eastward to the sea via two possible pathways: one is surface currents and the other one is turbidites along submarine canyons. The sedimentation rates of the terrigenous mud on the continental slope off eastern Taiwan range from 2.0 cm/kyr to 4.5 cm/kyr [*Shuford*, 1977, *Boggs et al.*, 1979; *Hung and Chung*, 1994]. These relatively low sedimentation rates are in contradiction with the high stream discharge and extremely high denudation rates in eastern Taiwan [*Li*, 1976, *Dadson et al.*, 2003], suggesting that much of the fine sediment discharged onto the shelf by eastern Taiwanese rivers must be transported beyond the inner slope by northward flowing Kuroshio surface current and thus are prevented from dispersing via eastward surface currents.

[24] Numerous submarine canyons running across the Luzon arc slope and submarine fan belt in the western half of the Huatung Basin are responsible for carrying sediment derived from the eastern part of Central Range and the Coastal Range to the Huatung Basin [*Liu et al.*, 2011]. The Huatung Basin, located just east of Taiwan, is an almost enclosed basin surrounded by the Yaeyama Ridge to the north, Gagua Ridge to the east, and the Luzon Arc to the south and west. The Gagua Ridge is a north-south trending, linear and continuous ridge, over 300 km long, 20 to 30 km wide, and rises 2 to 4 km above the adjacent sea floor, which isolates the Huatung Basin in the west from the main western Philippine Basin to the east. The sediments transported via submarine canyons from the Taiwan mountain belt are dammed by the Gagua Ridge, which is the main mechanism of sediment accumulation in the Huatung Basin. This results in sedimentation rates near the east coast of Taiwan and in the Huatung Basin [*Boggs et al.*, 1979; *Hung and Chung*, 1994] about ten times higher than those in western Philippine Basin (less than 1 cm/kyr, *Chen and Chung*, [1990]) on the east side of Gugua Ridge. This is supported by the thickness of the sediment cover of the Huatung Basin, which is about 500 m thicker than the western Philippine Basin [*Deschamps et al.*, 1998]. This suggests that the sediment eroded from the eastern Taiwan orogen is mainly trapped in Huatung Basin and cannot cross the Gagua Ridge to any significant extent into western Philippine Basin. Therefore, we conclude that the terrigenous signal in the sediment on Benham Rise does not originate from the eastern Taiwan orogen.

## 4.5. Is Asian Dust a Significant Sediment Source?

[25] It has been demonstrated that the Asian dust is transported eastward by the East Asian Monsoon [*Liu and Feng*, 1990; *Sun et al.*, 2001; *Liu et al.*, 2009] and prevailing westerlies across the marginal seas of the western Pacific even reaching the distant central North Pacific Ocean [*Duce et al.*, 1980; Rea, 1994; *Asahara et al.*, 1995]. Evidence from backward trajectories of air masses and dust observation analyses indicate that the eolian dust derived from Asian continent can also be transported southeastward to the South China Sea [*Hsu et al.*, 2008] and to Taiwan [*Liu and Feng*, 1990; *Liu et al.*, 2009]. Radiogenic Sr and Nd isotopic provide strong evidence that a significant fraction of the detrital sediments of the North Pacific are derived from Chinese loess (Central Province) and

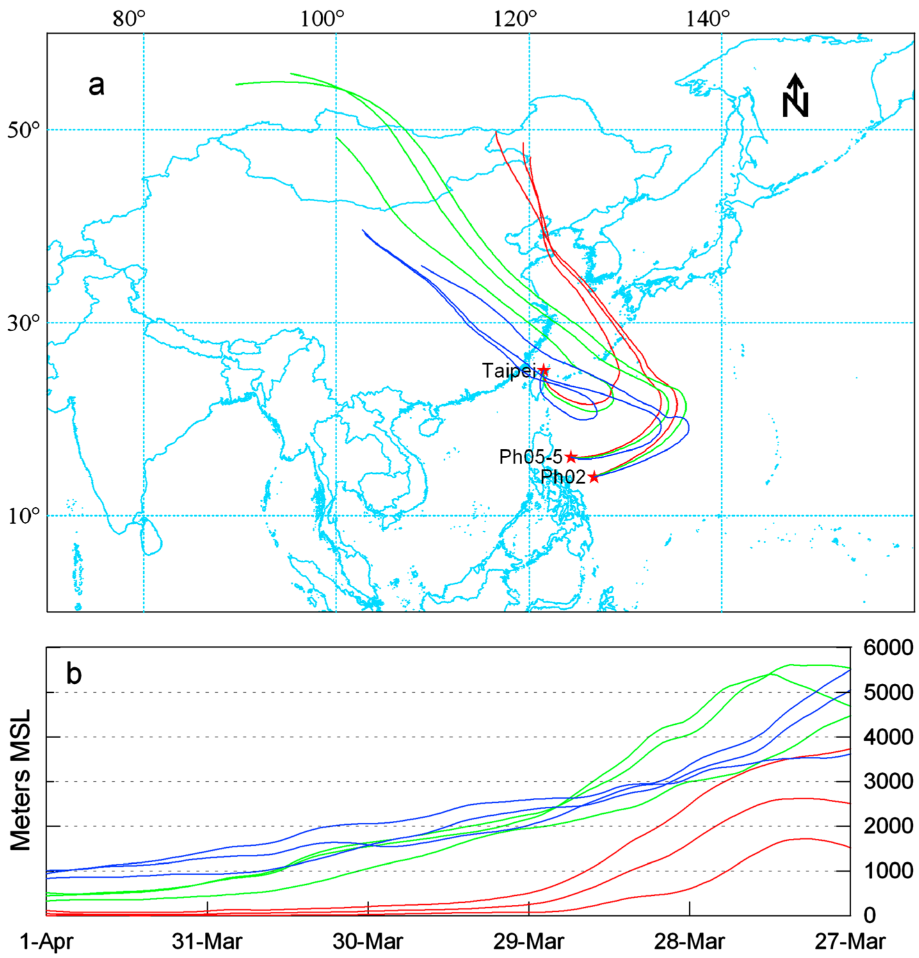


**Figure 5.** Trajectories of an example of a dust event from 27 March to 1 April 2006 and their (a) source locations and (b) vertical profiles.

relatively young crustal provinces around the Pacific Rim (Marginal Province) [*McLennan et al*., 1990; *Nakai et al*., 1993; *Jones et al*., 1994; 2000; *Weber et al*., 1996]. The sediment in the central North Pacific is less influenced by volcanic and island arc-derived material. Long distance transport makes the radiogenic Sr and Nd isotope composition of the detrital sediment in the central North Pacific very homogenous and potential sorting effects on the Sr isotope composition can be ignored as discussed previously. Given that our study area is located far from the Asian continent (more than 1500 km), it is reasonable to choose the homogenized sediment in the central North Pacific Ocean [*Pettke et al*., 2000] as the Asian dust end-member composition (The concentrations of Sr and Nd and isotopic compositions used here are [Sr] = 214 ppm, [Nd] = 38 ppm, $^{87}Sr/^{86}Sr = 0.71449$, and $^{143}Nd/^{144}Nd = 0.51217$ for average eolian dust in the North Pacific). The Luzon Islands are kept as the juvenile Sr and Nd isotope end-member [*Defant et al*., 1990]. The theoretical mixing curve of these two components is different from that of eastern Taiwan orogen and Luzon Island. The Nd isotopic ratio of the sediment derived Taiwan orogen is less radiogenic than that of dust in central North Pacific (Figure 4). The results also show that almost all the sediments on Benham Rise and in western Philippine Basin fit the mixing line very well. Because sorting during transport could have modified the Sr/Nd ratio and consequently the mixing trajectories, we evaluated the error envelope resulted from the grain size effect. According to *Feng et al*. [2009], different grain sizes result in about ±20% fluctuations in Nd/Sr ratio of eolian dust. Because we do not know the Nd/Sr ratios of the different grain size fractions of the Luzon arc sediments and eastern Taiwan sediments, we do not know the exact range of Nd/Sr ratios but would assume it is small given the prevailing rock types. We conservatively assumed ±20% variability in the Nd/Sr resulting from the sorting of the eolian dust, eastern Taiwan sediments, and for Luzon arc sediment and have calculated the error envelope of the mixing curve, respectively (Figure 4). The results show that the two mixing curves are still distinct, which supports that the detrital fraction in western Philippine Sea is a two component mixture between the Luzon Island and Asian dust. According to the mixing curve, the relative contribution of Asian dust to the detrital fraction on Benham Rise and the Philippine Sea basin is between 10% and 50% (Figure 4). These estimates have a maximum total uncertainty of about 20% (i.e., the above range is between 10% ± 2% and 50% ± 10%) as a function of the chosen end-member Nd and Sr isotope compositions and concentration. Results from a separate study indicate that the deposition rate of eolian dust in a long core sediment (MD06-3050) on Benham Rise has ranged between 0.1 and 0.9 g $cm^{-2}$ $kyr^{-1}$ (Jiang et al., unpublished data), which is similar to northwest Pacific [*Rea*, 1994], and hence supports that Asian dust is a significant detrital sediment source in the study area. To support this interpretation, we analyzed the backward trajectories of particle arrival height of 100, 500, and 1000 m above three sites, including the sites on Benham Rise (e.g., Ph05-5), western Philippine Basin (e.g., Ph02), and Taipei [*Liu et al.*, 2009] during the major dust event from 26 March to 1 April 2006 (Figure 5). The results show that all the three sites can be traced back to the same source area on the eastern Asian continent, which shows that the southeastward transported dust from the Asian continent influenced not only Taiwan [*Liu et al.*, 2009] but also the western Philippine Sea. This supports our conclusion that Asian dust contributes a significant part of the detrital fraction to the sediment of western Philippine Sea, which offers the potential to reconstruct changes in the climatic evolution of eastern Asia and compare this evolution to that of central China, which is the main dust source to the North Pacific.

[26] It may be argued that Asian continent material can be discharged to the Pacific Ocean by rivers and ocean currents. However, the geological background of the study area does not support such opinion. The sediments delivered by rivers are dominantly trapped in the marginal seas of China, such as the East China Sea and the Okinawa Trough [*Liu et al.*, 2007]. The obvious decrease of sedimentation rates from the East China Sea and Okinawa Trough (more than 50 mm/kyr, *Meng et al.*, [2009]) to western Philippine Basin (about 2.2–8.8 mm/kyr, *Huh et al.*, [1992]) and the northward transported western Pacific boundary current also indicates that it is difficult to transport the sediments delivered by rivers to pass through the wide East China Sea shelf, the deep back-arc basin (Okinawa Trough), and the Ryukyu Islands to the western Philippine Basin.

## 5. Conclusion

[27] The Sr and Nd isotopic signatures of detrital fraction of sediments from the western Pacific have been examined to test the applicability of distinguishing the provenance of deep-sea sediments. According to a two end-member mixing model, the mixing line of Asian dust and Luzon Islands is different from that of the eastern Taiwan orogen and Luzon Islands. Compared with eastern Taiwan orogen sediment, the Nd isotope ratio of Asian dust is significantly more radiogenic. By this mixing line, we can conclude that a significant fraction of evolved Sr and Nd signature in detrital fraction of the western Philippine Sea sediment are derived from Asian dust rather than from eastern Taiwan orogen. Our conclusion is supported by backward trajectories of the air masses and offers the potential for reconstructions of the climatic evolution of eastern Asia from sediments of the Philippine Basin.

## Acknowledgments

[28] The authors thank two reviewers, Y. Asahara and J. B. Mahoney, and editor L. Derry for detailed and constructive reviews and suggestion. We are grateful to J. Heinze for her support in the laboratory in Kiel. We also appreciate the help of C. Blanchet, P. Grasse, E. Hathorne, S. Kraft, M. Molina Kescher, A. Osborne, C. Teschner, J. Voigt, R. Stumpf, and M. Zieringer and for providing assistance in the course of the analytical work. The authors thank professor K-Y Wei of National Taiwan University for providing useful data for this paper. We appreciate the support from the Pilot Project of the National Knowledge Innovation Program of the Chinese Academy of Science grants KZCX2-YW-221 and the National Natural Science Foundation of China grants 40976026, 41230959, and 41106043. National Basic Research Program of China grants 2007 CB819503. The authors gratefully acknowledge the NOAA Air Resources Laboratory (ARL) for the provision of the HYSPLIT transport and dispersion model and/or READY website (http://ready.arl.noaa.gov) used in this publication.

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